Major factors controlling nitrous oxide emission and methane uptake from forest soil

ZHANG Xiu-jun
(*Shenyang College of Education, Shenyang 110015, P. R. China*)
XU Hui, CHEN Guan-xiong
(*Laboratory of Ecological Process of Trace Substance in Terrestrial Ecosystem, Institute of Applied Ecology, Chinese Academy of Sciences, Shenyang 110015, P. R. China*)

Abstract: Soil samples were taken from depth of 0-12 cm in virgin broad-leaved Korean pine mixed forest in Changbai Mountain in July 2000. The effects of temperature, soil water content, pH, NH_4^+ and NO_3^- on N_2O emission and CH_4 uptake of a forest soil were studied in laboratory by the method of orthogonal design. It was observed under laboratory conditions in this study that there were significant correlations between N_2O emission rate, CH_4 oxidation rate, soil pH and temperature. Nevertheless, N_2O emission rate also showed a significant positive correlation with CH_4 oxidation rate. The results suggested that pH and temperature were important factors controlling N_2O emission and CH_4 oxidation under this experiment conditions.

Key words: N_2O emission; CH_4 uptake; Orthogonal design; Forest soil
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Introduction

Nitrous oxide (N_2O) and Methane (CH_4) are two important greenhouse gases and also play an important role in photochemical reactions in atmosphere. The global warming potentials of N_2O and CH_4 are estimated at about 290 and 62 times that of carbon dioxide respectively. The concentrations of these gases have been increasing rapidly since the start of the industrial age, currently at rate of 0.25% and 1% per year respectively. It was reported that 70%-90% of these gases was of biogenic origin (Bouwman 1990). From the estimation of known global sources and sinks of N_2O made by IPCC (1996), the emission amount from temperate forests was 0.1-2 $Tg\cdot a^{-1}$. Total amount of atmospheric CH_4 consumed by aerobic soils ranged from 15 $Tg\cdot a^{-1}$ to 45 $Tg\cdot a^{-1}$, which is about 3% to 10% of the global emissions (Watson *et al.*1992). Forest ecosystem may function as a significant source for atmospheric N_2O and as a significant sink for atmospheric CH_4 within terrestrial ecosystems.

During the past more than ten years, lots of research have been conducted on N_2O emission and CH_4 uptake from different ecosystems, and the effects of various factors on N_2O emission or CH_4 uptake were studied, such as soil temperature, soil moisture, soil redox potential, soil pH, the kind of fertilizer used, and ecotype, etc. However, most of present points about these factors come from studies separately on N_2O emission or CH_4 uptake. Very few reports on interrelation between two gases' emission (consumption) can be found. Xu Hui *et al.* (1999) reported the trade-off relationship between N_2O emission and CH_4 consumption in forest soil. The relationships between N_2O emission and CH_4 uptake in rice paddy and upland field were also found respectively (Bai *et al.* 2000). Although N_2O emission and atmospheric CH_4 consumption are different processes in soils, it is possible that there are some common factors of controlling N_2O emission and CH_4 uptake. Up to now, there are very few investigations on N_2O emission and CH_4 uptake with forest soil. In this paper, effects of temperature, moisture, pH, NH_4^+ and NO_3^- on CH_4 oxidization and N_2O emission from a mixed broad-leaved Korean pine forest in Changbai Mountain were studied in laboratory. The objectives of this study were; (1) To try to find out what were the possible common controlling factors for N_2O emission and CH_4 uptake. (2) To make a further understanding of the interrelation between N_2O emission and CH_4 uptake.

Materials and methods

Experiment site

The sample plot was located in the virgin mixed broad-leaved Korean pine forest, at altitude 736 m in Changbai Mountain (42°24′N, 128°28′E). The annual mean precipitation is 700-800 mm. The soil is mountain dark brown forest soil. Soil samples were taken from a depth of 0 - 12 cm on July 1, 2000. Fresh soil was air-dried, sieved (2 mm). After air-dried, water content in the soil was 54.1%

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($g \cdot g^{-1}$ dry soil), soil NH_4^+-N and NO_3^--N contents were 7 and 17 ($\mu g \cdot g^{-1}$ dry soil) respectively and soil acidity in KCl was 5.15.

Experiment design

In orthogonal design, 5 factors and 5 levels were used to study the effects of these five factors on N_2O emission and CH_4 uptake in this experiment (Table 1), such as Temperature for 4, 9, 14, 19 and 24 °C; Water content for 60%, 85%, 110%, 135% and 160% ($g \cdot g^{-1}$ dry soil); Soil pH for 3.5, 4.5, 5.5, 6.5 and 7.5 (KCl $1 mol \cdot L^{-1}$); Soil NH_4^+-N and NO_3^--N content is 7, 17, 27, 37, 47 and 17, 27, 37, 47, 57 µg /g in dry soil respectively. The five levels were based on seasonal changes of the soil components. Three replicates were established for each combination.

Table 1. Factors and levels used for this study

Factors	Water content (%)	Temperature / °C	pH	NH_4^+ /mg·kg^{-1}	NO_3^- /mg·kg^{-1}
1	85	4	6.15	7	27
2	60	14	5.15	27	17
3	135	24	7.15	47	47
4	110	19	3.15	17	37
5	160	9	4.15	37	57

Notes: NH_4^+content means N content (g) in dry soil (kg); NO_3^- content means N content (g) in dry soil (kg); pH was measured in KCl (1mol/L).

Incubation procedure

The air-dried and sieved soil samples for 23 g (dry weight equivalent) were adjusted to the desired five pH-levels with NaOH 10 $mol \cdot L^{-1}$ or HCl 8 $mol \cdot L^{-1}$. The solutions containing NH_4Cl and KNO_3 were sprayed to the soil with a fine mist to create the desired water content, NH_4^+ and NO_3^-concentration. Each soil sample was mixed, and then was placed into a few 300-mL bottles. Each bottle was covered with a film for keeping fresh. The soil samples were equilibrated to a temperature of 15 °C for two days. After two days the film was lifted off the bottles. The bottles were sealed with rubber stoppers and were capped after they had been flowed with air for one minute, and then were incubated steady at temperature of 4, 9, 14, 19, 24 °C respectively. After initial sampling, the incubation lasted for 12 h for analyzing N_2O and CH_4 concentration in the headspace of the bottles by GC method.

Analysis of N_2O and CH_4 concentrations

N_2O concentrations were measured with Shimadzu GC-14A equipped with ECD. Detector, oven and injector temperature was 300, 60, and 100 °C, respectively. A Shimadzu GC-14B equipped with FID was used for CH_4 measurement. Detector, oven and injector temperature was 180, 140, and 100 °C, respectively.

Results and discussion

Effects of the five factors on N_2O emission rate

The results of the effects of the five factors on N_2O emission rate are shown in Table 2.

The result showed that the effects of temperature and pH on N_2O emission rate were significant (P<0.05 and P<0.01 respectively), but the effects of the other three factors on N_2O emission rate were not significant.

Table 2. Analysis of variance dependent variable of N_2O emission rate

Source	Sum of Square	*df*	Mean Square	*F* value
Water content	19.48	4	4.87	0.26
Temperature	2329.27	4	582.32	30.65**
pH	485.15	4	121.29	6.39*
NH_4^+	42.69	4	10.67	0.56
NO_3^-	48.65	4	12.16	0.64
Error	75.98	4	19.00	
Sum	3 001.22	24		

Notes: ** and * indicate significant at 1% and 5% levels respectively.

Multiple regression analysis of N_2O emission rate with the five factors

The positive correlations between N_2O emission, temperature and pH (P<0.01) were found (Table 3). But there were not significant relationships between N_2O emission rate and the other three factors under the experiment conditions.

Correlations between N_2O emission rate, temperature and pH

The relationships between N_2O emission rate, temperature and pH were found to be positive correlations (Table 3). The relationships were described as the following equation:

$$Y(N_2O)=1.333\,(T)+2.767\,(pH)-19.284 \qquad (1)$$

Where:

Y (N_2O) is the N_2O emission rate (ng $N_2O \cdot g^{-1} \cdot d^{-1}$), T is the temperature (°C).

When the temperature varied from 4°C to 24 °C, there was a positive relationship between N_2O emission rate and temperature (Table 2). The result indicated that the temperature variation was one of main factors controlling N_2O emission. In general, denitrification and nitrification of microorganism are known as two main N_2O production processes in the terrestrial ecosystem. The incubation study showed that Q_{10} values of nitrification and denitrification processes of the soil were 2.8 (6-28 °C) and 2.78 (6-28°C), respectively (Xu *et al.* 1999). A Q_{10} value (more than 2) indicated a significant effect of temperature on this biological process. Our result is consistent with their one. Therefore, the effect of temperature on N_2O emission in soil may be the result of temperature's effect on nitrification and denitrification processes.

There was also a positive correlation between N_2O emission rate and pH when the pH was at the range be-

tween 3.5 and 7.5 (Table 3). This result was also consistent with other studies. Paavolainen *et al.* (2000) reported that nitrification in ammonium-enriched soil suspensions was pH-dependant. Low pH led to decrease numbers of nitrifiers. Optimum pH of nitrification was about 7.0. In general, low pH not only can decrease the rate of denitrification, but also can increase the ratio of N_2O to N_2. The positive correlation between N_2O emission and pH found in this study indicated that nitrification might be a dominant process of N_2O production in the soil. The result indicated that temperature and pH were two important factors controlling N_2O emission in this experiment.

Table 3. Correlation coefficients between N_2O emission rate and the five factors

Item	NH_4^+	NO_3^-	Water content	Temperature	pH
N_2O emission rate	7.188E-02	4.546E-02	-2.120E-02	1.333**	2.767**

Notes: "**" indicates significant at 1% level.

Effects of the five factors on CH_4 oxidation rate in the soil

The results of the effects of the five factors on CH_4 oxidation rate are shown in Table 4.

Table 4. Analysis of variance dependent variable for CH_4 oxidation rate

Source	Sum of Square	*df*	Mean Square	*F* value
Water content	86.32	4	21.58	3.06
Temperature	219.14	4	54.78	7.75*
pH	268.08	4	67.02	9.48*
NH_4^+	13.51	4	3.38	0.48
NO_3^-	22.73	4	5.68	0.80
Error	28.25	4	7.06	
Sum	638.02	24		

Note: Level of significance, $^*P < 0.05$.

The result of incubation experiment revealed that the effects of temperature and pH on CH_4 oxidation rate were significant, but effects of the other three factors on CH_4 oxidation rate were not significant.

Multiple regression analysis of CH_4 oxidation rate and the five factors

CH_4 oxidation rate also showed significant positive correlations with temperature and pH in this experiment (Table 5). The relationships were presented as the equation (2):

$$Y(CH_4) = 0.27\ (T) + 1.813\ (\mathrm{pH}) - 5.728 \qquad (2)$$

Where:

$Y(CH_4)$ is the CH_4 oxidation rate ($ng \cdot g^{-1} \cdot d^{-1}$) in dry soil, T is the temperature (°C).

The result of multiple regression showed that CH_4 oxidation rate was affected by both soil pH and temperature (Table 5).

It was reported that soil temperature appeared to be an important controller of CH_4 consumption at low temperatures (-5 to 10 °C), but CH_4 consumption was independent of soil temperature between 10-20 °C (Castro 1995). Steinkamp *et al.* (2001) also reported at low soil temperature (<10 °C), temperature was a stronger modulator than soil moisture for CH_4 oxidation, but soil moisture was dominant factor controlling CH_4 oxidation when soil temperature was >10 °C. It was considered that a temperature threshold might exist, which was close to 10 °C. Our result was consistent with their studies. In our study, the correlation coefficients between CH_4 oxidation rate and temperature were 0.2 (P<0.05, when temperature was at the range between 4-14 °C) and 0.035 (P<0.05) (when temperatures were at the range between 14-24 °C) respectively. The temperature threshold of CH_4 oxidation rate in the soil may be about 14 °C. These results indicate that effects of soil temperature on CH_4 oxidation rate are dependent on the range of soil temperature.

Correlation analysis revealed that CH_4 oxidation rate was positively correlated with pH within a range of 3.5-7.5 (Table 5). The optimum pH for methane consumption was in the mesophilic range (Hutsch *et al.* 1994). Because pH value was lower than the optimum value for methane consumption in this experiment, CH_4 oxidation rate increased with the increasing pH. This result showed that optimum pH for methane consumption in the soil was about 7.0 .

Table 5. Correlation coefficient between CH_4 oxidation rate and the five factors

Item	NH_4^+	O_3^-	Water content	Temperature	pH
CH_4 oxidation rate	-4.22E-02	-3.4E-02	2.34E-02	0.27*	1.813**

Notes: ** and * indicate significant at 1% and 5% levels respectively.

Correlation between N_2O emission rate and CH_4 oxidation rate

Sitaula and Bakken (1993) reported that the rate of N_2O release was positively correlated with nitrification rate and the rate of CH_4 uptake was negatively correlated with nitrification rate in the incubation experiments with spruce forest soil. The reverse relation between N_2O and CH_4 fluxes from grassland soil was reported by Mosier (1991). The relationship was explained by the inhibition of NH_4^+ on CH_4 uptake (Mosier 1991). Xu Hui *et al.* (1999) also reported the trade-off relationship between N_2O emission and CH_4 consumption, and the effect of soil water content was used

for explaining this negative nonlinear correlation. Previously unreported results of this study is that there was a significant linear positive correlation between N_2O emission rate and CH_4 oxidation rate in our study (R^2=0.2702, P<0.01), (Fig.1).

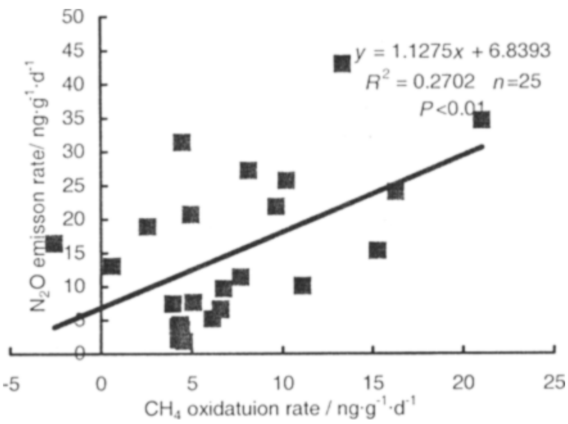

Fig. 1 Correlation of N_2O emission rate and CH_4 oxidation rate

By taking equation 1, 2 together into consideration, it seems possible to give a preliminary explanation for the mechanism of the positive correlation. Sitaula and Bakken (1993) indicated that nitrification and CH_4 oxidation were controlled by some common factors, which theoretically could be NH_4^+ availability. However, the measured NH_4^+ did not fit into this explanation, since the rates of N_2O emission and CH_4 oxidation were not correlated with NH_4^+ concentration respectively.

Although water content was considered as strong regulating factors on N_2O emission and CH_4 uptake respectively, It can not be considered as a common regulating factor for N_2O emission and CH_4 uptake, because no significant correlations between N_2O emission rate, CH_4 oxidation rate and water content were found in our study (Table 3 and Table 5). The factors that were found in N_2O-related equation and CH_4-related equation were soil pH and temperature. Therefore, the possible linkage between N_2O emission and CH_4 oxidation are soil pH and temperature. The positive correlation between N_2O emission rate and CH_4 oxidation rate was likely to be caused by variations of pH and temperature. This result indicates that pH and temperature may be common controlling factors for N_2O emission and CH_4 consumption in certain conditions. It is worthy of further investigating what relationship between N_2O emission and CH_4 consumption is when pH and temperature are fixed.

Conclusions

The results of Analysis of Variance and multiple regressions showed that there were significant correlations between N_2O emission rate, CH_4 oxidation rate, soil pH and temperature under certain lab conditions. The result indicated these two processes of N_2O emission and CH_4 oxidation were all controlled by pH and temperature. However, the significant positive correlation between N_2O emissions rate and CH_4 oxidation rate was also observed in this experiment.

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